Band structure engineering of type-II GaSb/GaAs quantum rings for intermediate band solar cells

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***Abstract*—Type-II quantum-confined heterostructures constitute a promising approach to realise highly efficient intermediate band solar cells (IBSCs), due to their long radiative lifetimes and the flexibility with which their electronic and optical properties can be engineered. To quantify the potential of type-II GaSb/GaAs quantum rings (QRs) for IBSC applications we undertake a theoretical investigation of their electronic properties. In these heterostructures the intermediate band is formed by bound hole states which are strongly localised within the QR. We demonstrate that the unconventional QR geometry provides a flexible platform to engineer the valence band structure, enabling optimum energy gaps – which maximise overall IBSC efficiency – to be obtained via structural optimisation. Our results emphasise that utilising QRs for IBSC applications requires careful control of QR morphology in epitaxial growth.**

I. Introduction

IBSCs constitute a promising approach to enhance photovoltaic efficiency, via introduction of an intermediate band (IB) into the band gap of a host matrix semiconductor. This allows for increased efficiency via two-step photon absorption (TSPA) – from the valence band (VB) to the IB, and from the IB to the conduction band (CB) – of photons having energies lower than the band gap of the host matrix semiconductor. An ideal IBSC has a maximum theoretical efficiency of 63.2%, significantly in excess of the 40.7% Shockley-Queisser limit for an ideal single-junction cell under concentrated illumination [1], [2].

Practical realisation of IBSCs is technically challenging. Firstly, the efficiency is limited by radiative loss of carriers excited by absorption into the IB. This represents a trade-off from the perspective of IBSC design, requiring suitably high absorption to excite carriers from the VB to the IB, but also that IB carriers have sufficiently long radiative lifetimes to enable TSPA to occur. Secondly, maximum IBSC efficiency is obtained, for a given host matrix band gap $E_g = E_{\rm CB} - E_{\rm VB}$, at specific intermediate gaps $E_{\rm L} = E_{\rm IB} - E_{\rm VB}$ and $E_{\rm H} = E_{\rm CB} - E_{\rm IB}$, but falls off rapidly in response to small shifts in $E_{\rm IB}$ [3]. This mandates growth of highly uniform heterostructures, having precisely controlled electronic properties. Thirdly, electrical isolation of the IB mandates the use of heterostructures providing a discrete density of states – i.e. three-dimensional confinement – which must be grown at sufficiently high densities to provide high optical absorption for practical applications.

Due to their intrinsically high radiative lifetimes, type-II heterostructures have attracted increasing attention as a suitable platform for the realisation of IBSCs [2]. In this work, we investigate the electronic properties of type-II GaSb/GaAs QRs. These type-II heterostructures, which provide confinement of holes but not of electrons, can be used to realise "hole-based" IBSCs in which the IB is formed by the QR hole ground state [4], [5]. The strain relaxation mechanism by which GaSb/GaAs quantum dots transform into QRs facilitates the growth of large QR stacks, providing high QR density to enable high optical absorption. We undertake an analysis of the electronic properties of these structures in response to changes in QR morphology, demonstrating (i) that QRs provide a flexible platform for band structure engineering, and hence IBSC optimisation, and (ii) that QRs having either low height h or large inner radius a_1 provide optimum IB energy $E_{\rm IB}$. Our calculations emphasise the importance of morphology in determining the suitability of QRs for IBSC applications.

II. Theoretical model

Our analysis of the electronic structure of cylindrical GaSb/GaAs QRs is based upon a multi-band **k·p** Hamiltonian. The strain fields in a calculational supercell – i.e. a GaSb QR and surrounding GaAs matrix – are computed using continuum elasticity theory, by minimising the total elastic energy of the entire supercell with respect to the displacement field. Using these strain fields we compute (to second order) the strain-induced piezoelectric polarisation, the electrostatic potential associated with which is included explicitly in our electronic structure calculations. The type-II GaSb/GaAs band alignment allows for confinement of holes, but not of electrons. To investigate the properties of the IB it is therefore sufficient to consider only the VB structure. On this basis, we employ a strain-dependent 6-band (Luttinger-Kohn + Bir-Pikus) **k·p** Hamiltonian for our electronic structure calculations, which describes the coupled heavy-hole (HH), light-hole (LH) and spin-split-off VBs. The calculations of the strain fields, piezoelectric potential and QR electronic structure are performed using a reciprocal space plane wave approach, as implemented in the S/Phi/nX software library [6]. The parameters used in our calculations are as recommended by Vurgaftman et al. [7]. All calculations are performed at temperature $T = 300$ K.

III. Results

For a given host matrix semiconductor band gap E_g the photovoltaic efficiency of an IBSC is a sensitive function of the IB energy $E_{\rm IB}$, and hence of the intermediate gaps $E_{\rm L}$ and $E_{\rm H}$. For a GaAs host matrix having $E_g = 1.42$ eV, a theoretical maximum efficiency $\approx 58\%$ is achieved for intermediate gaps $E_{\rm L} = 0.45$ eV and $E_{\rm H} = E_g - E_{\rm L} = 0.97$ eV [3]. For a hole-based IBSC utilising GaSb QRs embedded in a GaAs matrix, optimum efficiency can therefore be realised by engineering the QR VB structure so that the (predominantely HH-like) hole ground state lies 0.45 eV above the GaAs VB edge in energy. We begin with typical QR dimensions – inner and outer

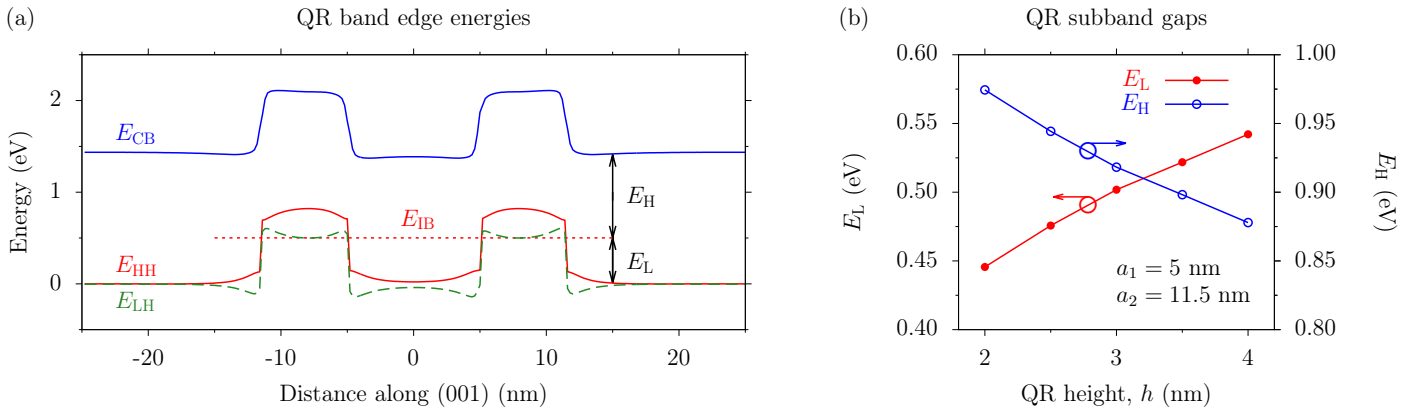

Fig. 1. (a) Linescans along the (100) direction of the CB (solid blue line), HH (solid red line) and LH (dashed green line) band edge energies for a (001)-oriented GaSb/GaAs QR having inner and outer radii $a_1 = 5$ nm and $a_2 = 11.5$ nm, and height $h = 3$ nm. The horizontal dotted red line denotes the calculated energy E_{IB} of the IB – i.e. the predominantly HH-like QR hole ground state. (b) Calculated variation of the VB to IB and IB to CB intermediate gaps E_L (closed red circles) and E_H (open blue circles) with QR height h, for GaSb/GaAs QRs having inner and outer radii $a_1 = 5$ nm and $a_2 = 11.5$ nm.

radii $a_1 = 5$ nm and $a_2 = 11.5$ nm, and height $h = 3$ nm – based on experimental characterisation [4], [5], and compute the extent to which the VB structure is modified in response to the changes in QR morphology. On the basis of these computed trends we then draw general conclusions regarding ideal QR morphology from the perspective of IBSC optimisation.

Figure 1(a) shows the calculated CB (solid blue line), HH (solid red line) and LH (dashed green line) band edge energies along the (100) direction in the plane of a GaSb/GaAs QR having typical dimensions. We firstly note the type-II band alignment, providing a large ≈ 0.8 eV VB offset and hence strong confinement of HH-like VB eigenstates. Strain relaxation produces small residual tensile strain in the centre of the QR, slightly lowering the CB edge energy. However, we calculate that this minor reduction in CB edge energy, combined with the CB offset in a strained GaSb/GaAs QR, is insufficient to provide appreciable electron confinement. The compressive strain in the QR produces a large VB offset for HH valence states, with the QR hole ground state – the energy E_{IB} of which is denoted by a dotted red line – calculated to be predominantly HH-like. Figure 1(b) tracks the change in energy of this state – quantified via the intermediate gaps E_L (closed red circles) and E_H (open blue circles) – as a function of QR height h. Our calculations indicate that, for typical QR inner and outer radii, optimum intermediate gaps $E_L = 0.45$ eV and $E_H = 0.97$ eV are achieved in shorter QRs having $h \approx 2$ nm. Our calculations therefore suggest that GaSb/GaAs QRs represent a promising platform to realise IBSCs, since optimum intermediate gaps can be obtained in structures compatible with established epitaxial growth.

IV. Conclusion

We have undertaken a theoretical investigation of the electronic properties of GaSb/GaAs QRs, and demonstrated that these heterostructures provide significant flexibility for IBSC design and optimisation. The type-II GaSb/GaAs band alignment can be expected to mitigate radiative losses of carriers from the IB, while the energy of the IB formed by the confined QR hole ground state can be readily engineered to its optimum value via relatively small changes in QR morphology. For typical QR radii – $a_1 = 5$ nm and $a_2 = 11.5$ nm – our calculations indicate that an optimum VB to IB (IB to CB) intermediate gap of 0.45 eV (0.97 eV) can be obtained for QRs of height $h \approx 2$ nm. Alternatively, for taller QRs having fixed outer radius, optimum IB energy can be engineered by increasing the inner radius a_1. QRs grown to these specifications have a theoretical maximum efficiency $\approx 58\%$, close to the theoretical limit for ideal IBSCs. Given the sensitivity of IBSC efficiency to E_{IB}, our analysis suggests that careful control of QR morphology provides a viable route to realising heterostructures suitable for IBSC applications.

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